

Effect of the Compounding Ingredients on the Thermal Conductivity of Synthetic Rubber

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The thermal conductivity of synthetic rubber, where the amount of compounding ingredients is changed, is obtained by the comparative method with a steady heat flow. The thermal conductivity increases with the increase of the amount of vulcanizing agent, reinforcing agent and other agents, and slightly decreases with temperature rise when the amount of the compounding ingredients in the synthetic rubber is not large. Generally, since the reinforcing agent like carbon black etc. is considered to exist as discontinuous phase in the continuous phase of pure rubber, the thermal conductivity of the synthetic rubber can be calculated by Eq. (1) which shows the thermal conductivity for the mixtures of heterogeneous materials.

I. Introduction

Synthetic rubber is used in large numbers in the industries, and its physical properties have been investigated closely. However, in these properties, it is considered that the effect of compounding ingredients on the thermal conductivity of the synthetic rubber has not been satisfactory determined. In practical use, the thermal conductivity of the synthetic rubber comes into a considerable important problem because that the internal heat generation according to high speed revolution for the tire of motor car and vibration for the rubber isolator attached to machines, and etc. are discussed. From this, it is desired generally that the thermal conductivity of the synthetic rubber is good in the many cases.

In this paper, the effects of carbon black, vulcanizing agent, reinforcing agents, fillers and etc. on the thermal conductivity of the synthetic rubber will be discussed on the basis of the experimental results.

2. Experimental Apparatus and Method

The thermal conductivity of the synthetic rubber was measured by the comparative

method with a steady heat flow. Figure 1 shows an outline of the experimental apparatus. A square shaped experimental sample and standard sample are placed together, these are enclosed by two water jackets, and silicon grease is applied to hold in close contact between them. In order to prevent heat loss from the edges, the circumference of the samples is surrounded by sub-heater. For the standard sample, a Teflon plate of 50×50 mm size and 5 mm thickness with a known thermal conductivity (the thermal conductivity $\lambda = 0.160 + 0.0002 t$ kcal/mh $^{\circ}$ C, where t is average temperature) is used.

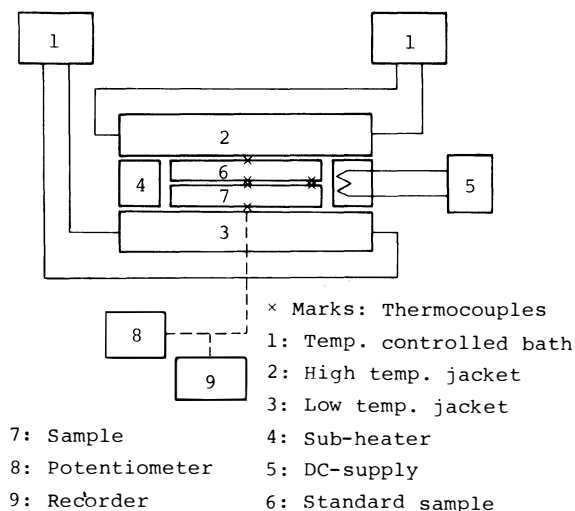


Fig. 1. An outline of experimental apparatus

C-C thermocouples with a diameter of 0.08 mm are used for temperature measuring, each one of which is attached to front and back surfaces near the center of the standard sample and the experimental sample, and each two of them are attached at the edges, respectively. The voltage of the thermocouples is measured to an accuracy of 0.01° C with a potentiometer.

For the experiments, when constant temperature waters with temperature control to within $\pm 0.02^{\circ}$ C are circulated through the high and low temperature jackets, the temperature difference between both jackets is held to about 15° C, and the apparatus is allowed to attain a steady state by a recorder, the temperature gradients for the experimental sample and the standard sample are measured.

In addition, for investigation of the experimental accuracy for present apparatus, the comparison of thermal conductivity is carried out for two samples of the same material and same shape, which are placed together, and experimental error within 2 % was confirmed when the sub-heater was used properly.

3. Experimental Samples

The samples for the synthetic rubber of the present experiments are shown from No. 1 to No. 11 in Tables 1 ~ 3, in which the weight parts of the compounding ingredients against 100 parts of pure rubber are shown. Table 1 shows four samples of Butadiene rubber (BR) in which the amount of sulfur is changed, and Table 2 shows three samples of Acrylonitrile-Butadiene rubber (NBR) in which the amount of carbon

black is changed. Table 3 shows four samples consisting of Isoprene rubber (IR) as main body, in which the type and the amount of the compounding ingredients are changed.

Table 1. The weight parts of compounding ingredients against 100 parts of pure rubber for Butadiene rubber.

No.	1	2	3	4
BR	100	100	100	100
ZnO	5	5	5	5
St. acid	1.5	1.5	1.5	1.5
HAF	40	40	40	40
Acc. CZ	0.8	0.8	0.8	0.8
Sulfur	1.0	1.5	2.0	2.5

Vulcanization. 160 °C, 20 min

Table 2. The weight parts of compounding ingredients against 100 parts of pure rubber for Acrylonitrile-Butadiene rubber.

No.	5	6	7
NBR	100	100	100
ZnO	5	5	5
St. acid	1	1	1
Acc. DM	1	1	1
Acc. D	1	1	1
Sulfur	1.5	1.5	1.5
FEF	20	40	60

Vulcanization. 155 °C, 20 min

Table 3. The weight parts of compounding ingredients against 100 parts of pure rubber for practical Isoprene rubber.

No. 8		No. 9		No. 10		No. 11	
IR	70	IR	60	IR	100	IR	70
EPDM	30	1778J	40	ZnO	2	NR	30
ZnO	5	ZnO	5	St-Zn	1	ZnO	5
St. acid	1	St. acid	2	Sulfur	2	St. acid	2
Sulfur	2.5	Sulfur	2.5	TC-talc	60	Sulfur	2.5
SRF-LM	50	Pre-CaCO ₃	90	TiO ₂	5	Pre-CaCO ₃	70
Calmos	20	D-clay	30	Acc. BG	0.5	TC-talc	10
LPO	10	HAF	10	Acc. TS	0.3	Acc. DM	1.5
Acc. DM	1.5	LPO	20			Acc. DT	0.5
Acc. D	0.3	YS-resin	2			Paraffin	0.5
Acc. BM	3	Acc. CZ	0.8				
		Acc. D	0.5				
		Acc. SL	0.6				

In these tables, the sulfur is vulcanizing agent, HAF, FEF and SRF-LM are the type of carbon black which serves as reinforcing agent, and D-clay, Calmos, TC-talc, Pre-CaCO₃ serve as extending agent and filler. Acc.-CZ, BM, SL, BG, etc. are the type of vulcanizing accelerator agent, ZnO, St-Zn, stearic acid are activator, and lubricant (LPO), coloring agent (TiO₂), etc. are contained. In Table 3, EPDM, 1778J and NR are Ethylene-Propylene rubber, Styrene-Butadiene rubber and natural rubber, respectively.

In the experiments, the samples of 50 × 50 mm in size and 3 ~ 5 mm in thickness were used.

4. Results and Considerations

Figure 2 shows the relation between the thermal conductivity at 25 °C and the amount of sulfur for Butadiene rubber. The thermal conductivities increase linearly as shown by a straight line in the figure as the amount of sulfur is increased. In general,

it is known that the thermal conductivity for amorphous polymeric materials has a tendency to increase in the temperature above the glass transition temperature T_g as the crosslinking density of molecules is increased, and that there is a linear relation between the thermal conductivity and the crosslinking density¹⁾. The Butadiene rubber for the present experiments is also an amorphous polymeric material, and its glass

transition temperature is about -70°C . As sulfur which is a vulcanizing agent increases the crosslinking density in Butadiene rubber, it is seen that the tendency of the increase for the thermal conductivity in Fig. 2 coincides with the above fact.

Figure 3 shows the relation between the thermal conductivity at 25°C and the amount of carbon black for Acrylonitrile-Butadiene rubber. The thermal conductivities have a tendency to increase with increasing amount of carbon black. Usually, the pure rubber and the carbon black in rubber materials do not chemically combine, and the carbon black is considered to exist as discontinuous phase in the continuous phase of the pure rubber.

The carbon black is the greater part of spheres with $40 \sim 80 \text{ m}\mu$ in size, and its micro-structure is considered to be amorphous²⁾. It is known that the thermal conductivity of amorphous carbon is about $1.7 \text{ kcal/mh}^\circ\text{C}$ at 20°C ³⁾ and increases with the increase of the temperature. On the other hand, the thermal conductivity of the pure rubber for NBR can be estimated as $0.2 \text{ kcal/mh}^\circ\text{C}$ from the extrapolation of the experimental values in Fig. 3.

In general, for the thermal conductivity of heterogeneous mixtures, the following equation⁴⁾ has been known to be applicable

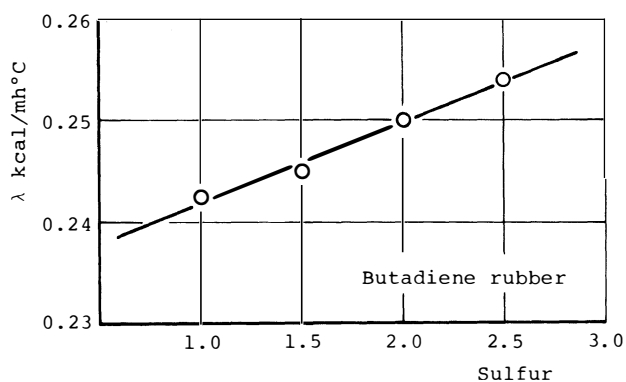


Fig. 2. Relation between the thermal conductivity and the weight parts of sulfur against 100 parts of pure rubber.

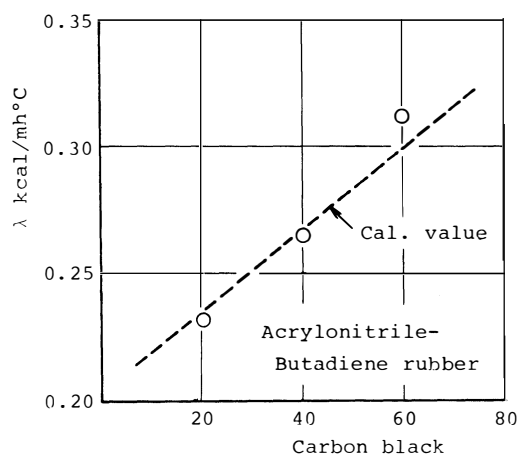


Fig. 3. Relation between the thermal conductivity and the weight parts of carbon black against 100 parts of pure rubber.

$$\lambda = \lambda_1 \left[\frac{\lambda_2 + (n-1) \lambda_1 - (n-1) V_2 (\lambda_1 - \lambda_2)}{\lambda_2 + (n-1) \lambda_1 + V_2 (\lambda_1 - \lambda_2)} \right] \quad (1)$$

where, λ_1 and λ_2 are the thermal conductivity of the continuous and discontinuous phases, respectively, V_2 is the volume ratio for the discontinuous phase, and n is 3 when the discontinuous phase consists of sphere particles. The thermal conductivities of NBR calculated by Eq. (1) are shown by a broken line in Fig. 3. The experimental and calculated values almost agree within the error of several percent. Therefore, Eq. (1) may be applied to such case that the rubber is considered to be the mixtures of pure rubber and carbon black.

In Fig. 4, the temperature dependence of the thermal conductivities for NBR samples is shown. The thermal conductivities as approximated by the straight lines in the figure have a tendency to decrease slightly with the temperature rise. For the amorphous polymeric materials, it is known that in the lower temperature than the glass transition temperature T_g the molecular chains are congealed and the thermal motion of molecules is limited, but that the thermal motion becomes active with temperature rise. From this, the thermal conductivity in the temperature under the glass transition temperature increases with temperature rise⁵⁾.

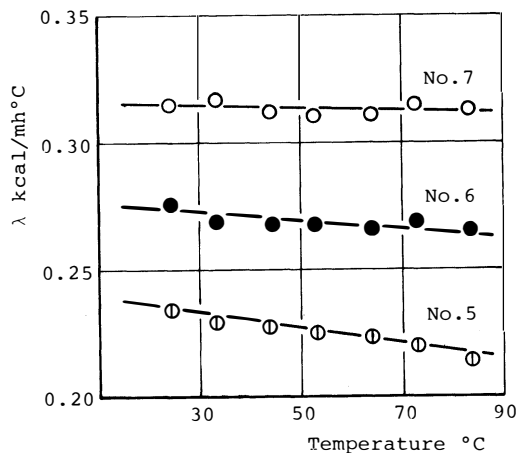


Fig. 4. Temperature dependence of the thermal conductivity for Acrylonitrile-Butadiene rubber.

In the higher temperature than T_g , the constraining force and the combining force of molecules decrease with temperature rise, so that the heat flow through the molecules will decrease and the thermal conductivity also decreases. Therefore, the thermal conductivity for pure rubber of NBR is considered to decrease with temperature rise. On the contrary, as the thermal conductivity of carbon black increases with temperature rise, the decrease rate for the thermal conductivity of NBR is considered to decrease gradually with increasing carbon black, as shown by the straight lines in Fig. 4.

Figure 5 shows the temperature dependence of the thermal conductivities for the practical rubber in Table 3. These rubbers contain the fillers like D-clay, Calmos, TC-talc and Pre- CaCO_3 . In general, it is considered that the fillers do not chemically combine with the pure rubber in the same as carbon black²⁾. The sum of the weight percentage for the carbon black and fillers are 41 %, 57 %, 35 % and 42 % to the

samples of No. 8 ~ No. 11, respectively. When the weight percentages are converted into the volume percentages, they become about 20 %, 27 %, 13 % and 21 %, respectively. In Fig. 5, a tendency that the thermal conductivities increase with increasing volume percentage of the carbon black and fillers is shown.

The sample of No. 8 contains Calmos as fillers in addition to carbon black. Calmos is a commercial name of the type of precipitated calcium carbonate and its thermal conductivity is about $1.9 \text{ kcal/mh}^\circ\text{C}$ at 20°C ³⁾. Therefore, it is considered that Calmos has nearly same effect as carbon black to the thermal conductivity of the rubber.

In the sample of No. 9, Pre- CaCO_3 is also the type of precipitated calcium carbonate and is considered to have the almost same effect as Calmos to the thermal conductivity. D-clay consists of the aluminum silicate as the main body and its thermal conductivity is $6.0 \text{ kcal/mh}^\circ\text{C}$ ⁶⁾. In the sample of No. 9, the volume percentage of D-clay is about 5.6 % and the volume percentage of the sum of Pre- CaCO_3 and carbon black is about 21.4 %. The sum of those is about 27 % and is largest in IR samples. Therefore, the thermal conductivity of this sample shows the largest value in IR samples.

The sample of No. 10 is called white rubber and contains TC-talc as reinforcing agent in place of the carbon black. TC-talc consisting of magnesium silicate as the main body has the thermal conductivity of about $5.2 \text{ kcal/mh}^\circ\text{C}$ ⁶⁾. Its influence to the thermal conductivity of the synthetic rubber is considered to be fairly larger than that of the carbon black. However, as the volume percentage of TC-talc is about 13 % and is smallest in IR samples, the thermal conductivity of this sample will show the smallest value.

The sample of No. 11 is called brown rubber and contains Pre- CaCO_3 and TC-talc as reinforcing agent. The sum of their volume percentage is nearly same rate as the sample of No. 8. However, the thermal conductivity of TC-talc is larger than that of the carbon black, so that the thermal conductivity of this sample will be larger than that of the sample of No. 8.

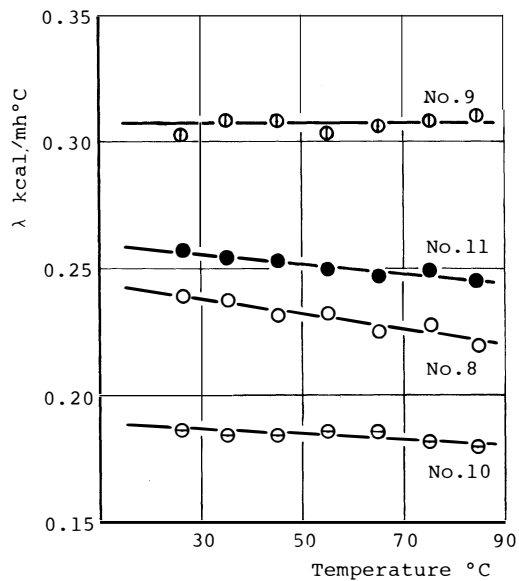


Fig. 5. Temperature dependence of the thermal conductivity of practical Isoprene rubber.

5. Conclusions

Experiments were done for the thermal conductivity of synthetic rubber, and the following results were obtained.

(1) The thermal conductivity of synthetic rubber increases slightly with increasing amount of vulcanizing agent.

(2) The thermal conductivity of synthetic rubber increases with increasing amount of carbon black, and the good agreement between the experimental values and calculated values by Eq. (1) is obtained.

(3) In practical rubber, the thermal conductivity has a tendency to increase with the increase of the volume percentage of reinforcing agents and fillers.

(4) The thermal conductivity of synthetic rubber decreases slightly with temperature rise.

Acknowledgment

The authors thank Mr. Iwao Wada of Nippon Zeon Company for his help with the manufacture of the samples etc..

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The main part of this paper is expressed to preprint of Japan Soc. Mech. Engrs. (No. 767-1), at Toyama, September 1976.

(Received. October 31. 1978)